

issues” as previously decided by the Board. In reply, that is not correct. The claims contain the limitations of Claims 6 **and** 7. No appealed claim contained all of these limitations. Nor is it relevant that Applicants argued during the appeal that each of Claims 6 and 7 stands or falls separately. Based on the Board’s opinion, it does not appear that the Board considered the patentability of Claims 6 and 7 separately from that of Claim 1. In addition, as previously pointed out, and as pointed out again below, a significant fact found by the Board in affirming the rejection was that the amount of stabilizer (c) recited in the claims overlapped, in effect, the amount of biuretizing agent disclosed by Wagner et al (Decision at 8-9). In the present claims, the maximum amount of stabilizer is less than the minimum amount of biuretizing agent necessary, as disclosed by Wagner et al and by Hennig et al, *infra*. Another fact found by the Board in reaching its decision was that the data in the specification was not explained (Decision at 11). The significance of this data was explained in the previous response, and is repeated below.

Rejected Claims 10, 11 and 13-15 are additionally not governed by *res judicata* because none of the appealed claims limited the Markush group for stabilizer (c) in Claim 1. In other words, there was never an issue before the Board with regard to the patentability of individual members of the Markush group.

For all the above reasons, it is respectfully requested that the rejection on grounds of *res judicata* be withdrawn.

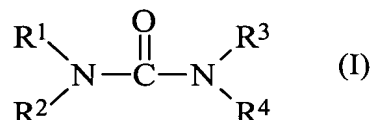
The rejection of Claims 1-5, 8-11 and 13-17 as unpatentable under 35 U.S.C. §103(a) over US 4,192,936 or US 4,152,350, each to Möhring et al in view of US 3,903,127 or US 3,976,622, each to Wagner et al, and US 3,367,956 to Hennig et al, is respectfully traversed.<sup>1</sup>

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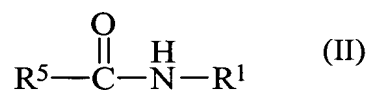
<sup>1</sup>The disclosures of each of the Möhring et al patents, and the Wagner et al patents, respectively, are identical. Thus, we refer herein to US 4,152,350 of Möhring et al and US

The invention relates to a process for the preparation of a polyisocyanate which contains one or more biuret groups by reacting

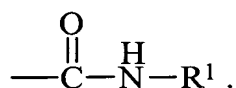
- a) an aliphatic or cycloaliphatic isocyanate containing two or more isocyanate groups (isocyanate (a)) with
- b) 0.5 to 20 mol% based on the isocyanate groups in (a) of a tertiary alcohol or a mixture of water and a tertiary alcohol (biuretizing agent (b)) at from 100 to 250°C, which comprises carrying out the reaction in the presence
- c) from 0.01 to 2.0 mol% based on the isocyanate groups in (a) of a stabilizer (c) selected from the group consisting of urea, ammonia, biuret, a urea derivative of the formula I



in which R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup> and R<sup>4</sup> are hydrogen, C<sub>1</sub> to C<sub>10</sub> alkyl or C<sub>6</sub> to C<sub>10</sub> aryl, or a carboxamide of the formula II



in which R<sup>5</sup> is C<sub>1</sub> to C<sub>12</sub> alkyl which is unsubstituted or in which 1, 2 or 3 hydrogen atoms are replaced by a radical



As discussed in the specification, the biuret-containing polyisocyanate prepared by the known processes from tertiary alcohols and isocyanates leave much to be desired, since they are too dark in color for many applications and, in particular after prolonged storage, still include considerable quantities of readily volatile monomeric isocyanates.

Applicants have discovered an economic process by whose use it is possible to prepare biuret-containing polyisocyanates which are pale in color and whose contents of volatile isocyanates, particularly after prolonged storage, is low.

An essential feature of the claimed invention is the presence of a stabilizer (c) which is present in now numerically-recited amounts and selected from a Markush group of compounds. It is due to the presence of such component (c) as a **catalyst, not as a biuretizing agent**, in place of other known catalysts, such as those disclosed by Wagner et al at column 6, lines 30 to 42, that, unexpectedly, an improved product is obtained, as demonstrated by the comparative evidence set forth in the specification for the products according to the Examples of the invention in Table 1 at page 9, compared to the results of the Comparative Examples in Table 2 at page 10. A copy of Tables 1 and 2 is **attached herewith**.

As is evident from the results set forth in these Tables, it is apparent that the products obtained by the claimed process evince significantly lower color numbers as well as their monomer content being significantly and materially lower after 21 days of storage.

As disclosed at page 2, lines 1-5 of the specification, it is probable that during the reaction in a first step a urethane is formed which in a second step is decomposed into an amine, CO<sub>2</sub> and an olefin. In a third step this amine forms urea with additional isocyanate and finally this urea forms biuret with additional isocyanate.

Since during the process the amine is formed in situ, no amine has to be introduced

initially. The reaction products of the claimed process are practically allophanate-free biurets even though a tertiary alcohol is used as a reagent therein.

Möhring et al discloses a process for the preparation of polyisocyanates containing biuret groups, which is disclosed as substantially eliminating disadvantages of prior art processes, such as the release of monomeric isocyanate reactant in the course of prolonged storage (column 2, line 58 - column 3, line 2). Möhring et al's invention involves the use of certain mixtures of alcohols, primary amines and optionally water as biuretizing agents (column 3, lines 3-6). Among the advantages disclosed by Möhring et al is resulting polyisocyanate mixtures having a high proportion of biuret polyisocyanates having formulae (I) and (II) therein, and allophanate polyisocyanates having a formula (III) therein (column 3, lines 37-44). While Möhring et al disclose primary, secondary and tertiary alcohols, the preferred alcohols are monohydric primary alcohols (column 5, lines 59-62). However, as noted by both the Board and the Examiner, Example 6 of Möhring et al employs a tertiary butanol, and appears to result in the formation of no allophanate groups.

Wagner et al, which Applicants contend is still the closest prior art, even though the Board has found that Möhring et al is, has been relied on for their disclosure of some of the presently-recited stabilizers, and more specifically N,N'-disubstituted ureas, as biuretizing agents (paragraph bridging columns 5 and 6, and particularly column 6, line 7ff). Hennig et al has been relied on for its disclosure of substituted ureas as biureting agents (paragraph bridging columns 1 and 2).

The Board's rationale is that it would have been obvious to add any of the biuretizing agents of Wagner et al or Hennig et al to the already-present biuretizing agents of Möhring et al, that the claims on appeal did not exclude additional biuretizing agents, and that such additional biuretizing agents "can include the 'catalytic' and any excess amount of 'stabilizer'

which is itself a recognized biuretizing agent" (Decision at 8).

In reply, the present claims now require that the stabilizer (c) be present in a maximum amount of 2.0 mol.% based on the isocyanate groups. Note that the Board found that Wagner et al discloses a molar ratio of diisocyanate to biuretizing agent, extending up to about 40:1, i.e., about 2.5 mol.% (Decision at 8-9). Thus, the maximum amount of stabilizer of the present claims is less than the minimum amount of biuretizing agent necessary, as disclosed by Wagner et al. In addition, while Hennig et al discloses reacting at least three moles of diisocyanate with a substituted urea (column 1, lines 56-58), none of the examples therein employs a diisocyanate to substituted urea ratio greater than 10:1. In other words, the substituted urea is present in an amount of at least 10 mol% based on the diisocyanate.

The Board found that the meaning of the above-discussed data at pages 9 and 10 of the specification is not explained (Decision at 11). In reply, as described in the specification at page 2, lines 25-28 and 37-39, the monomers used are highly toxic and should therefore be as low as possible in the final product. Indeed, this knowledge is confirmed by Möhring et al. The specification discloses at page 11, line 35ff that the monomer content after 21 days means how much monomers are released during storage for 21 days at 50°C. It should be clear from comparing the amounts both directly after preparation (0d) and after 21 days of storage (21d), that the change for the Examples is considerably less than for the Comparative Examples.

The significance of low color number is described in the specification at page 2, lines 31-32, and page 7, lines 40-42; given the fact that the polyisocyanates are mainly used in the paint industry, one skilled in the art would appreciate that they should be substantially colorless.

As further evidence that the presently-recited stabilizer is not simply a biuretizing

agent, Examples 9, 11, 12 and 13 can be compared. These examples are all identical, except for the amount of stabilizer, i.e., urea, used, as described in the specification at page 8, lines 13-14 and 21-23. Data from Examples 9 and 11-13 have been abstracted from the above-discussed Table 1, with addition of a column "Amount of Stabilizer", in the table below:

Ex.	Biuretizing agent (b)	Stabilizer	Amount of Stabilizer	Temp.	NCO content	Viscosity	Color number
9	tBuOH: water 1:1	UR	0.2	180	22.2	5450	4
11	tBuOH: water 1:1	UR	0.4	180	22.0	6120	12
12	tBuOH: water 1:1	UR	0.6	180	21.3	11,560	18
13	tBuOH: water 1:1	UR	1.0	180	20.8	18,200	22

As the above table shows, the greater the amount of stabilizer employed, the greater is the viscosity and the greater is the color number. The increase of viscosity is presumably due to the formation of higher biurets which is catalyzed by the stabilizer. Thus, the stabilizer acts as a catalyst for biuret formation, but not as a biuretizing agent. The increase of the monomer content within 21 days is nearly the same for these examples (0.12 - 0.17 wt.%). See Table 1. This data shows that it is even more advantageous to use less stabilizer.

Further comparing Examples 5 and 6 with Comparative Example 12, shows how the color number and the increase of monomers during storage, i.e., the difference between the value after 0 days and 21 days, becomes better using the stabilizers of the present invention:

Ex.	Biuretizing agent (b)	Stabilizer	Temp.	NCO content	Viscosity	Color number	Increase of Monomers
5	tBuOH: water 19:1	Eth UR	180	22.7	2200	12	0.30
6	tBuOH: water 19:1	DM UR	180	22.7	2280	15	0.30
Comp. Ex. 12	tBuOH: water 19:1	-	180	22.7	2090	32	0.49

It can be seen from the above-discussed data that using urea derivatives **does** affect color number and monomer increase advantageously. The influence of the stabilizer on color number and monomer content is neither disclosed nor suggested by any of the applied prior art. Indeed, given the rationale of the Board, no significant difference should have been expected in Examples 5 and 6, and Comparative Example 12, if the stabilizer acts as a biuretizing agent, given the fact that the biuretizing agent (b) is present in an amount of 14 mol%, while the stabilizer (c) is present in a much smaller amount, i.e., 0.2 mol%, as described in the specification at page 8, lines 12-14.

Claims limited to a stabilizer (c) component other than the particular urea derivatives disclosed by Wagner et al or Hennig et al are separately patentable, since none of the prior art discloses or suggests these stabilizer (c) components as biuretizing agents or stabilizers.

In the Office Action, the Examiner maintains his position that at least some of the members of the stabilizer (c) Markush group are disclosed as biuretizing agents, and it would thus have been obvious to combine known biuretizing agents absent a showing of unexpected results. In reply, the above-discussed comparative data shows that the stabilizer (c) has an effect on color number and monomer increase not suggested by the prior art.

The Examiner apparently finds that lightness in color would be expected, noting that

Hennig et al disclose that their biuret polyisocyanates are light in color. In reply, as discussed above, Hennig et al use significantly more substituted urea compound to make their biuret polyisocyanates than the maximum amount for the presently-recited stabilizer (c). More significantly, Hennig et al disclose in their Example 1 a "golden yellow solution;" Example 2 discloses a "light yellow mixture." In the presently-disclosed examples, a Hazen color number of 22 or less is obtained. See Table 1. As is well-known, a color number of 15 Hazen or less represents a coloration that is not visible to the naked eye and can be measured only with optical instruments. It is not likely that Hennig et al's Hazen color numbers are even close to 22. The light yellow mixture of Example 2 can be guessed as having a Hazen number of 50-100; the golden yellow solution probably has a Hazen number of several hundred.

Indeed, none of the applied prior art employs a compound that overlaps the compounds of stabilizer (c) in an amount within the recited amounts of the present claims. While the Examiner states, in paragraph 8 of the Office Action, that the Board agreed with the Examiner's findings with regard to the amount of stabilizer of the present claims compared to the amounts disclosed in Wagner et al, it is clear, as discussed above with regard to the rejection on grounds of *res judicata*, that the amounts recited in **all** the present claims differ from that of Wagner et al.

Regarding the comparative data of record, the Examiner finds that it does not "rise to the level of being unexpected." The Examiner's only basis for this is Hennig et al which, as discussed above, employ urea derivatives in a significantly greater amount than the maximum amount recited for the presently recited stabilizer (c). Nor does the Examiner say anything with regard to the increase of monomers.

In addition, and even more significantly, Hennig et al disclose that their biuret



polyisocyanates cannot be obtained by reacting compounds containing hydroxyl groups (column 3, lines 25-31). Thus, if anything, Hennig et al teach away from the presently-claimed invention, which requires a compounds containing hydroxyl groups as the biuretizing agent.

Claims 10, 11, 13, 16 and 17 are each separately patentable. The only biuretizing agents of Wagner et al or Hennig et al that overlap with the compounds within the terms of stabilizer (c) of Claim 1 are certain N, N'-disubstituted ureas. None of the stabilizer compounds of Claims 10, 11, 13, 16 and 17 are disclosed or suggested by the applied prior art as biuretizing agents.

Claim 10, which limits the stabilizer to urea *per se*, is separately patentable for an additional reason. Applicants have discovered that of all of the stabilizers of the present invention, urea provides the best results, both in the improvement of color as well as monomer content. This superiority is demonstrated by comparing Examples 3-6. Data for these examples has been extracted from Table 1, discussed *supra*, and reproduced below:

Ex.	Biuretizing agent	Stabilizer	Temp.	Color Number	Monomer Content [0d]	Monomer Content [21d]
3	tBuOH:water 19:1	UR	170	7	0.08	0.22
4	tBuOH:water 19:1	UR	190	10	0.10	0.21
5	tBuOH:water 19:1	Eth UR	180	12	0.15	0.45
6	tBuOH:water 19:1	DM UR	180	15	0.13	0.43

As described in the specification at page 11, UR means urea, Eth UR means ethylene urea, and DM UR means N,N'-dimethyl urea.

Although the temperature of Examples 3 and 4 is higher and lower, respectively, than that of Examples 5 and 6, it can be easily seen that both examples using urea, i.e., Examples 3 and 4, show better results with regard to color number as well as monomer content compared to the use of the substituted ureas of Examples 5 and 6. These results could not have been predicted by any of the applied prior art.

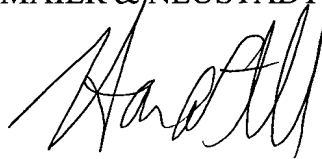
While Wagner et al disclose an embodiment wherein formamide or N-substituted formamides are used as reactive, viscosity-reducing additives, in which biuret polyisocyanates which contain formyl groups are obtained (column 9, line 7 ff), the entire disclosure related thereto in Wagner et al concerns the formation of formyl groups, whereby precursors other than formamide, such as formic acid, can be used. There is no disclosure or suggestion to use other amides such as acetamide, which is recited in present Claim 17. The Examiner's citation of *In re Grabiak*, 226 USPQ 870 (Fed. Cir. 1985) is inapposite. Indeed, *Grabiak* supports Applicants' position. In *Grabiak*, the Court found that the presence of oxygen in a compound did not render an analogous compound containing a sulfur atom *prima facie* obvious.

For all the above reasons, it is respectfully requested that the rejection over prior art be withdrawn.

Applicants gratefully acknowledge the Examiner's indication of allowability of the subject matter of Claims 12 and 18. Nevertheless, Applicants respectfully submit that all of the presently-pending claims are now in condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

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Table 1

Ex.	Biuretizing agent (b)	Stabilizer (c)	Temp. [°C]	NCO content [% by wt.]	Viscosity [mPa.s]	C N [Hazen]	Monomer content 0 d [% by wt.]	Monomer content 21 d [% by wt.]
1	tert-Butanol (tBuOH)	UR	180	22.0	4350	5	0.15	0.25
2	tBuOH	Eth UR	180	22.7	2290	10	0.20	0.41
3	tBuOH:water 19:1	UR	170	22.4	3340	7	0.08	0.22
4	tBuOH:water 19:1	UR	190	22.0	6030	10	0.10	0.21
5	tBuOH:water 19:1	Eth UR	180	22.7	2200	12	0.15	0.45
6	tBuOH:water 19:1	DM UR	180	22.7	2280	15	0.13	0.43
7	tBuOH:water 4.6:1	UR	180	22.2	5550	5	0.11	0.23
8	tBuOH:water 1.8:1	UR	180	22.0	6480	2	0.13	0.28
9	tBuOH:water 1:1	UR	180	22.2	5450	4	0.14	0.31
10	tBuOH:water 0.27:1	UR	180	21.4	12,600	10	0.14	0.28
11	tBuOH:water 1:1	UR	180	22.0	6120	12	0.12	0.27
12	tBuOH:water 1:1	UR	180	21.3	11,560	18	0.12	0.29
13	tBuOH:water 1:1	UR	180	20.8	18,200	22	0.13	0.25
14	tBuOH:water 19:1	Biuret	180	22.0	3860	15	0.14	0.27
15	tBuOH:water 19:1	Acetamide	180	22.6	3020		0.17	0.31
16	tBuOH:water 19:1	Samid	180	22.5	3000		0.14	0.34
17	tBuOH:water 19:1	Ammonia	180	22.0	2340	28	0.21	0.50

Table 2

Comp. Ex.	Biuretizing agent (b)	Acidic catalysts	Temp. [°C]	NCO content [% by wt.]	Viscosity [mPa·s]	C N [Hazen]	Monomer content 0 d [% by wt.]	Monomer content 21 d [% by wt.]
1	tert-Butanol (tBuOH)	BF <sub>3</sub>	150	22.9	2550	206	0.09	0.69
2	tBuOH	PTSS	150	21.7	5400	350	0.05	0.48
3	tBuOH	DEHP	180	22.0	4840	42	0.07	0.42
4	tBuOH	EHA	180	22.0	4660	38	0.09	0.42
5	tBuOH	HAC	180	22.1	4330	55	0.08	0.40
6	tBuOH	-	180	22.9	2130	44	0.09	0.53
7	tBuOH:water 19:1	PTSS	180	22.0	5550	371	0.11	0.91
8	tBuOH:water 19:1	PTSS	150	21.8	5360	256	0.03	0.49
9	tBuOH:water 19:1	DEHP	180	22.4	3800	32	0.10	0.53
10	tBuOH:water 19:1	EHA	180	22.4	3650	10	0.15	0.63
11	tBuOH:water 19:1	ClAc	180	22.3	3970	56	0.14	0.53
12	tBuOH:water 19:1	-	180	22.7	2090	32	0.12	0.61